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MAGNETIC PROPERTIES OF $REPb_2$(RE: LIGHT RARE-EARTH ELEMENT) COMPOUNDS

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Abstract- Systematic studies on the crystal structure and magnetic properties of light rare earth element(RE) compounds with Pb, $REPb_2$, have been carried out. Their crystal structure has been identified to be a $MoSi_2$-type. The values of the effective magnetic moment for $CePb_2$, $PrPb_2$ and $NdPb_2$ are respectively very close to the theoretical values of RE^{3+}. These three compounds are antiferromagnetic and exhibit a metamagnetic transition. The magnitude of the Néel temperature is proportional to two-thirds of the de Gennes factor. The magnetic entropy change for $NdPb_2$ is contrast to the value for $CePb_2$ heavy-fermion compound, comparable to the theoretical value. The magnetic contribution to the temperature dependence of resistivity for $PrPb_2$ is given by a form of $-lnT$ in a wide temperature range, implying the Kondo system in analogy with $CePb_2$.

I. INTRODUCTION

The study of magnetic properties for rare earth element compounds with Pb has been mainly focussed on compounds with a $AuCu_3$-type structure[1]. The antiferromagnetic magnetic ordering in these compounds occurs at rather low temperatures. Especially, the magnetic properties of $CePb_3$ have been investigated extensively because of the coexistence of the long-range magnetic order and the heavy-fermion behavior[2]. The compounds composed of the rare earth element and an element X with the composition ratio 1:2 are the largest family[3]. However, no systematic studies on their crystal structure and magnetic properties for the compounds with Pb have been made. Recently, the existence of $CePb_2$ has been confirmed and its heavy-fermion behaviors have been compared with those of $CePb_3$[4].

In the present study, the crystal structure and magnetic properties for $REPb_2$ compounds(RE = light rare earth element such as La, Ce, Pr and Nd) have been investigated. High-field measurements have been made because the low temperature antiffero-magnetic ordering would be correlated with a meta-magnetic transition.

II. EXPERIMENTAL

The starting materials were 99.9 % pure La, Ce, Pr and Nd and 99.999 % pure Pb. The alloying was made by arc-melting in an argon gas atmosphere purified with a Ti getter. The ingots were turned over and remelted five times to ensure homogenization. The powder X-ray diffraction was carried out using $CuK\alpha$ radiation. In order to prevent the powder samples from oxidizing, they were protected from the atmosphere by coating with Vaseline.

The magnetization up to 55 kOe and the

temperature dependence of the DC magnetic susceptibility from 2 to 300 K were measured with a SQUID magnetometer. The low-temperature specific heat from 1.8 to 15 K was measured by a conventional heat pulse method with a mechanical heat switch in an adiabatic cell and a 1 K pot. The samples were fixed with nylon lines in a chamber evacuated to about 10^{-7} Torr to obtain thermal isolation. The temperature dependence of electrical resistivity was measured by a conventional DC four-probe method.

III. RESULTS AND DISCUSSION

The structural analysis was made by X-ray diffraction. The $REPb_2$ compounds have been ascertained to have a body centered tetragonal $MoSi_2$-type structure(space group *I4/mmm*). This structure has also been confirmed in the compounds with the heavy rare earth element such as Gd, Tb or Dy. Furthermore, a clear lanthanoid contraction has been observed. Representative diffraction patterns have been presented elsewhere[4]. The crystal structure of a $PrPb_2$ compound has been reported to be a $HfGa_2$-type structure[5], inconsistent with the present result. However, one should notice that the (019) peak of the $HfGa_2$-type structure is very close to the (111) peak of pure Pb, which is often induced by selective oxidization of the rare earth element in Pb compounds.

Figure 1 shows the temperature dependence of magnetic susceptibility for $CePb_2$, $PrPb_2$ and $NdPb_2$. These curves exhibit a small peak or an inflection point. In addition, they show no magnetic cooling effect. These behaviors are characteristic of antiferromagnets. Shown in Fig. 2 is the Néel temperature T_N vs the de Gennes factor G. With increase in the value of G, the Néel temperature determined from the temperature dependence of magnetic susceptibility becomes higher. It has been pointed out that T_N for various heavy rare earth alloys

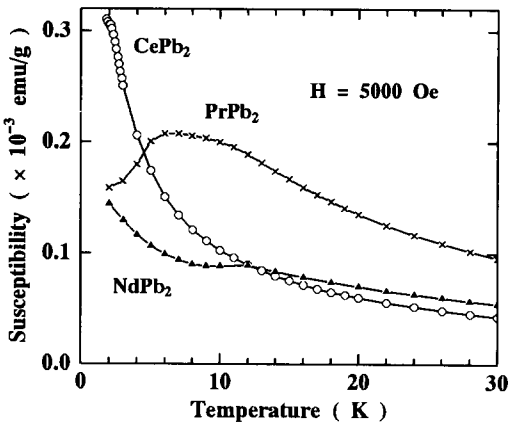

Fig. 1 Temperature dependence of the magnetic susceptibility of $CePb_2$, $PrPb_2$ and $NdPb_2$.

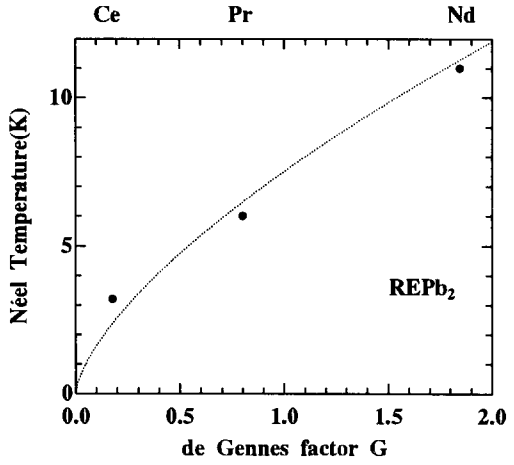

Fig. 2 The Néel temperature T_N vs the de Gennes factor G for three compounds.

with La, Y or Sc is proportional to $G^{2/3}$[6]. Even in the present light rare earth element compounds with Pb, this relation is held as seen from Fig. 2. The dotted line in the figure represents the fitted curve.

Figure 3 shows the magnetization curves for $CePb_2$ and $PrPb_2$. Since their Néel temperature is

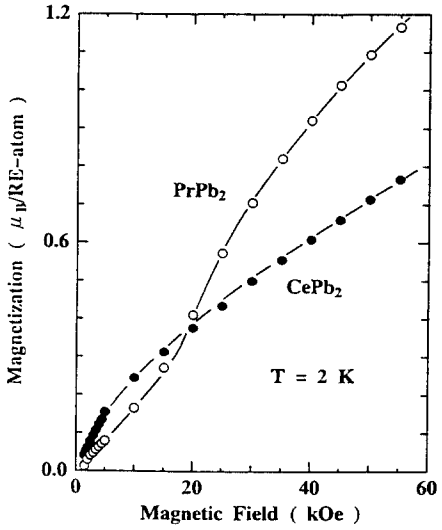

Fig. 3 Magnetization curves up to 55 kOe at 2 K for $CePb_2$ and $PrPb_2$.

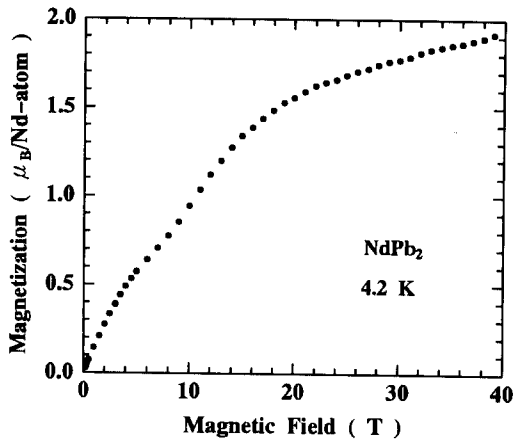

Fig.4 Magnetization curve measured up to very high fields at 4.2 K for $NdPb_2$.

very low as seen from Fig. 2, the measurements have been made at 2 K. The field dependence of magnetization is not monotonic. Namely, $CePb_2$ has two magnetic transition temperatures at very low temperatures[4]. Its magnetization curve is convex upwards, suggesting the occurrence of a metamagnetic transition at a very low magnetic field and/or a canted antiferromagnetic structure. On the other hand, $PrPb_2$ exhibits a linear increase up to 15 kOe and a marked increase in the magnetization occurs above 15 kOe due to the metamagnetic transition.

The magnetization curve measured at 4.2 K in pulsed magnetic fields for $NdPb_2$ is shown in Fig. 4. A marked increase in the magnetization is confirmed in the vicinity of 90 kOe. Furthermore, a non-linear field dependence in lower magnetic fields is observed in a similar manner as $CePb_2$. This behavior may be correlated with a canted antiferromagnetic spin structure. The neutron diffraction study on the spin structures and the magnetization measurements up to much higher fields would provide useful information.

The Néel temperature of $NdPb_2$ is higher than that of $CePb_2$ and $PrPb_2$ as seen from Fig. 2. Therefore, the higher the Néel temperature, the higher the metamagnetic transition field.

Figure 5 shows the temperature dependence of the inverse magnetic susceptibility χ^{-1} for three compounds. At high temperatures, the following Curie-Weiss law is set up

$$\chi = \chi_0 + C/(T - \theta_P) \qquad (1),$$

where χ_0, C and θ_P are the temperature independent susceptibility, the Curie constant and the paramagnetic Curie temperature, respectively. From the value of C, the effective magnetic moment μ_{eff} is obtained.

The magnetic data for the $CePb_2$, $PrPb_2$ and $NdPb_2$ compounds are summarized in Table 1. The experimental values of the effective magnetic moment μ_{eff} for these compounds are respectively almost the same as those theoretical values of RE^{3+}. The paramagnetic Curie temperature θ_P and the Néel temperature T_N obtained from Figs. 5 and 1, respectively, are also given in the same table.

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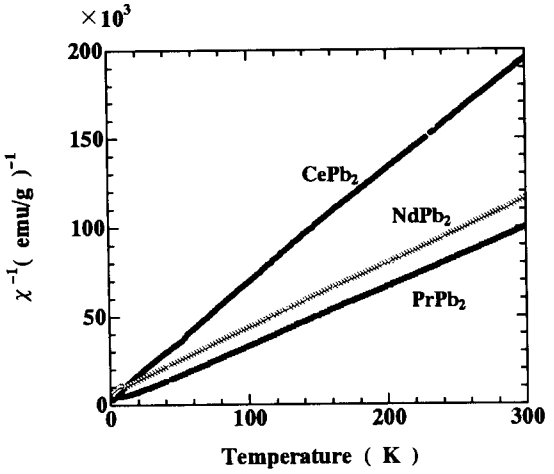

Fig. 5 Temperature dependence of the inverse magnetic susceptibility of $CePb_2$, $PrPb_2$ and $NdPb_2$.

Table 1 Effective magnetic moment μ_{eff}, paramagnetic Curie temperature θ_P, the Néel temperature T_N for $CePb_2$, $PrPb_2$ and $NdPb_2$.

Compound	$\mu(\mu_B)$	θ_P(K)	T_N(K)
$CePb_2$	2.54	-3.9	3.6
$PrPb_2$	3.70	-2.4	6.0
$NdPb_2$	3.44	-17.8	11.0

The low-temperature specific heat data for the $NdPb_2$ is given in Fig. 6, together with that for $LaPb_2$ as a reference compound. The latter exhibits a λ-type specific peak at the Néel temperature. The difference between the result for the antiferromagnetic $NdPb_2$ and that for the paramagnetic $LaPb_2$ gives the magnetic contribution, that is,

$$C_{mag} = C_{total} - (\gamma T + \beta T^3) \qquad (2),$$

where γ and β are the electronic and lattice specific heats, respectively. The magnetic entropy change is estimated by integrating the C_{mag}/T vs T curve. The experimental value for $NdPb_2$ is not so different from the theoretical value. It is noteworthy that the experimental value for $CePb_2$ is about 68 % of $Rln2$, implying that 32 % of 4f electrons in $CePb_2$ become non-magnetic because of itinerancy[4]. The lattice specific heat coefficient β is related to the Debye temperature Θ_D by the following expression:

$$\beta = (12/5)\pi^4 R(1/\Theta_D)^3 \qquad (3),$$

where R is the gas constant. The obtained value of Θ_D is 125 K, being similar to that of $CePb_2$[4].

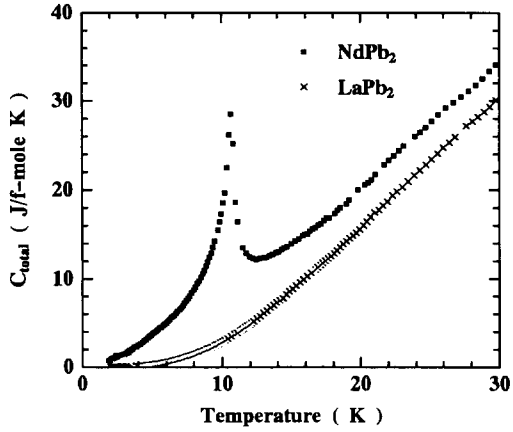

Fig. 6 Temperature dependence of the specific heat for $NdPb_2$ and $LaPb_2$.

Figure 7 shows the temperature dependence of the electrical resistivity of the magnetic contribution ρ_{mag} to the resistivity for $CePb_2$ and $PrPb_2$. The ordinate indicates the relative magnitude subtracted from the result for the paramagnetic $LaPb_2$. In high temperature ranges, it has been pointed out that ρ_{mag} for $CePb_2$ is in proportion to $-lnT$ with regard to the Kondo system[4]. The deviation from the logarithmic temperature dependence at low temperatures would be explained by the reduction in scattering due to the crystal-field splitting[7] and/or the development of the coherence between the Kondo sites[8]. It is

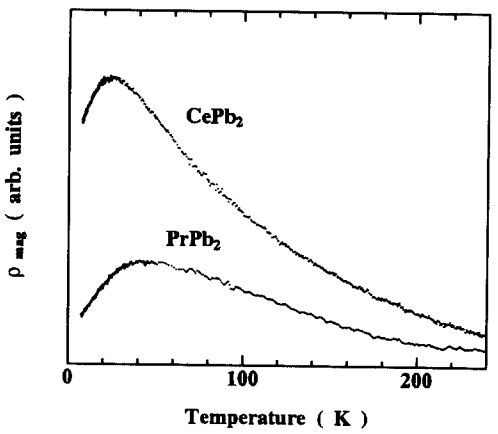

Fig.7 Temperature dependence of the magnetic contribution to the resistivity ρ_{mag} for $CePb_2$ and $PrPb_2$.

interesting to note that the temperature dependence of $PrPb_2$ is similar to that of $CePb_2$ heavy-fermion compound as seen from Fig. 7. That is, the temperature dependence of ρ_{mag} for $PrPb_2$ is also proportional to $-lnT$ in a wide temperature range. Detailed magnetic, specific heat and electrical investigations are necessary for further clarification.

IV. CONCLUSION

Crystal structure and magnetic properties of $REPb_2$(RE: light rare earth element such as La, Ce, Pr and Nd) compounds have been investigated. Main results are summarized as follows.

(1) Four $REPb_2$ compounds composed of La, Ce, Pr and Nd have a $MoSi_2$-type structure.

(2) These compounds, except $LaPb_2$, are antiferromagnetic and their Néel temperature is proportional to two-thirds of the de Gennes factor.

(3) The metamagnetic transition is caused by applying magnetic field. The higher the Néel temperature, the higher the critical field of the metamagnetic transition.

(4) The effective magnetic moments for $CePb_2$, $PrPb_2$ and $NdPb_2$ are respectively nearly equivalent to the theoretical values of E^{3+}.

(5) The magnetic entropy change for $NdPb_2$ is comparable to the value expected theoretically in contrast to the value for $CePb_2$ heavy-fermion compound.

(6) The magnetic contribution to the resistivity for $PrPb_2$ shows a negative logarithmic temperature dependence in a wide temperature range.

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